

1 **Prevalence of organic gunshot residues in police vehicles**

2
3 Anne-Laure Gassner, Céline Weyermann

4 School of Criminal Sciences, University of Lausanne, Switzerland

5 6 **Abstract**

7
8 The present study investigated the organic gunshot residue (OGSR) background level of police vehicles
9 in Switzerland. Specimens from 64 vehicles belonging to two regional police services were collected
10 and analysed by LC-MS in positive mode. The driver's and back seats were sampled separately to
11 monitor potential differences between locations and to assess the risks of a suspect being contaminated
12 by OGSR during transportation to a police station.

13 The results showed that most of the 64 vehicles were uncontaminated (44 driver's seats and 38 back
14 seats respectively). Up to six of the seven targeted compounds were detected in a single sample, once
15 on a driver's seat and twice on back seats. The contamination frequency generally decreased as the
16 number of compounds detected together increased. The amounts detected were in the low ng range and
17 less than amounts generally detected just after discharge on a shooter. Our data indicated that detecting
18 a combination of four or more compounds on a police vehicle seat appears to be a relatively rare
19 occurrence. The background contamination observed was most probably due to secondary transfer from
20 police officers (e.g. through recent participation in a shooting session or firearm manipulation) or from
21 firearms stored in the vehicles. The present results might be used as a recommendation to minimize
22 contact of a suspect with contaminated surfaces if OGSR is implemented in routine work in parallel to
23 IGSR analysis.

24
25 Keywords: Forensic science, firearm discharge residue, background, contamination, LC-MS

27 **1. INTRODUCTION**

28 Gunshot residues (GSR) are one of the forensically relevant traces produced by the discharge of a
29 firearm. The residues consist of vapour and particulate matter expelled mainly from the muzzle, but also
30 from other firearm openings, that might deposit onto the target, the shooter, potential bystanders and
31 objects close to the firearm [1, 2]. The production and transfer of GSR depend on a number of factors
32 such as the type of firearm, the ammunition, the number of shots fired, the properties of the recipient
33 material and the environmental conditions [3]. While in casework GSR are frequently used to estimate
34 the shooting distance or distinguish entrance and exit wounds, they also help in assessing the potential
35 involvement of an individual in a shooting incident [4, 5]. Inorganic GSR (IGSR) mainly originate from
36 the primer and other metallic parts of the firearm and ammunition (i.e. barrel, bullet and cartridge case),
37 whereas organic GSR (OGSR) are produced by incomplete combustion of the propellant [4-6]. While
38 IGSR are routinely analysed in forensic laboratories using scanning electron microscopy coupled to
39 energy dispersive X-ray spectroscopy (SEM-EDX) [1, 4], OGSR analysis is still rarely applied in
40 casework. This may be explained by the absence of a standardised protocol (collection, extraction and
41 analysis), as several analytical methods have been proposed and investigated for the analysis of
42 propellants and OGSR without one outperforming the others in all particulars. Spectroscopic techniques,
43 such as Raman [7-9], Fourier transformed infrared spectroscopy (FTIR) [10, 11] or ion mobility
44 spectrometry (IMS) [12-14] detect OGSR based on spectral information, without formal compound
45 identification. While these methods are non-destructive, their sensitivity remains limited (IMS) or is yet
46 to be demonstrated on real specimens (Raman, FTIR). On the contrary, bulk analytical techniques such
47 as micellar electrokinetic capillary electrophoresis (MEKC) [15-18], gas chromatography (GC) [19-21]
48 or liquid chromatography coupled to mass spectrometry (LC-MS) [22-27], separate and identify the
49 compounds, can be very sensitive, but involve the dissolution of the specimen.

50 Studies showed that GSR are lost relatively quickly from the hands, even without washing, resulting in
51 very low amounts still remaining on a shooter's hands a few hours after discharge [1]. Thus, very
52 sensitive analytical techniques are required. Recent OGSR forensic studies utilizing LC-MS analysis
53 have demonstrated the detection of OGSR on the hands of a shooter up to four hours after discharge
54 [28] and highlighted secondary transfer in several scenarios [29, 30]. Currently, OGSR can be detected
55 at the sub-picogram level, due to major improvements in MS sensitivities in the last decade. Moreover,
56 new technical developments are expected and should further enhance detection capabilities using LC-
57 MS, increasing the potential of this technique for the analysis of OGSR. However, sensitivity
58 improvements generally lead to increased background signal, requiring careful interpretation of the
59 results and evaluation of the various activities that can produce such traces.

60 The interpretation of GSR evidence requires background or prevalence studies in relevant populations,
61 which are case- and country-specific. GSR prevalence can depend on occupation, living area
62 (city/countryside, known firearm violence), and firearm possession (legal and illegal) but also on the
63 population type, e.g. individuals vs objects (clothing, vehicles, public places). For example, the

64 probability of finding GSR on the hands of a police officer who carries a service weapon and regularly
65 practices shooting might be higher than on a citizen with absolutely no contact with firearms.
66 Consequently, the evidentiary value of a GSR trace will vary accordingly with the case circumstances
67 and the explanation provided by the defense [31]. Background studies in a police environment might
68 also play a role in police management. Indeed, monitoring the GSR background would highlight
69 potential risks of secondary transfer from police officers and premises, thus helping in establishing
70 procedures to avoid such transfer to individuals arrested, transferred in police vehicles and detained in
71 police facilities.

72 Various background studies have been conducted, targeting IGSR and/or OGSR (Table 1). However,
73 their number remains relatively limited. Other types of studies aiming at reconstructing events of a
74 shooting case have also been reported. For example, a simulation of shooting was carried out to
75 quantitate GSR contamination of a car's interior surfaces when a firearm was discharged within a car,
76 showing a significant amount of characteristic IGSR particles on the window headliner and dashboard
77 [32].

DRAFT

78 Table 1: Summary of background/prevalence studies targeting IGSR and/or OGSR

Reference	GSR type	Population type	Population size	Country	Surface sampled	Analytical technique	Main results
Gialamas et al, 1995 [33]	IGSR	Police	43	USA	Hands	SEM-EDX	<ul style="list-style-type: none"> 3 specimens with one PbBaSb particle in a population of 43 non-shooting police officers
Berk et al, 2007 [34]	IGSR	Police	201	USA	Vehicles and detention facilities	SEM-EDX	<ul style="list-style-type: none"> total of 56 PbBaSb particles found in 23 specimens two vehicles with one particle 54 particles recovered from detention facilities with a maximum of seven particles collected from a table surface and restraining bars
Lindsay et al, 2011 [35]	IGSR	Firearm manufacture employees	13	Canada	Hands	SEM-EDX	<ul style="list-style-type: none"> PbBaSb particles found on nine of the employees no more than two characteristic particles found on the hands of the five individuals who had no direct contact with firearms for the other four employees: number of particles from nine to 424
Gerard et al, 2012 [36]	IGSR	Police	66 police officers 28 civilians working in police environment 18 vehicles	Canada	Hands, clothes, equipment and vehicles	SEM-EDX	<ul style="list-style-type: none"> at least one PbBaSb particle on the hands of 60% of patrol and plainclothes officers and on 24% of their equipment no IGSR particles found on the 28 civilians working in a police environment 2 of the 18 vehicles sampled had one characteristic GSR particle
Brozek-Mucha, 2014 [37]	IGSR	Civilian & police	50 shooters 100 non-shooters	Poland	Hands	SEM-EDX	<ul style="list-style-type: none"> one PbBaSb particle detected among individuals who had no contact with firearms numerous particles found among shooters showing a strong correlation with the time elapsed since the last shooting session
Hannigan et al, 2015 [38]	IGSR	Arrested people	100	Ireland	Upper body garments	SEM-EDX	<ul style="list-style-type: none"> 98% of the specimens collected from the cuffs negative up to two PbBaSb particles detected on two garments
Cook, 2016 [39]	IGSR	Police	33	Australia	Hands	SEM-EDX	<ul style="list-style-type: none"> 28 officers with PbBaSb particles on their hands, with an average of 64 such particles
Lucas et al, 2016 [40]	IGSR	Civilian	289	Australia	Hands	SEM-EDX	<ul style="list-style-type: none"> overall prevalence of 0.3% for characteristic PbBaSb particles, 8% for PbSb and about 7% for single Pb, Ba or Sb particles
Comanescu et al, 2019 [41]	IGSR	Civilian	50	USA	Vehicles	Graphite Furnace Atomic Absorption	<ul style="list-style-type: none"> no positive specimen
Lucas et al, 2019 [42]	IGSR	Police	76	Australia	Hands	SEM-EDX	<ul style="list-style-type: none"> 7.9% of the officers returned at least one characteristic PbBaSb particle 75% of the officers had at least one consistent particle (in average < 5)

Northrop, 2001 [15]	OGSR	Civilian	100	USA	Hands	MEKC	<ul style="list-style-type: none"> no positive specimen
Bell and Seitzinger, 2016 [43]	OGSR	Civilian	73	USA	Hands	IMS	<ul style="list-style-type: none"> less than 5% of positive specimens
Ali et al, 2016 [44]	IGSR & OGSR	Police	70	USA	Police stations	SEM-EDX & LC-MS	<ul style="list-style-type: none"> one characteristic IGSR particle detected (interview desk) ethylcentralite quantified in two specimens
Hofstetter et al, 2017 [45]	OGSR	Civilian and police	27 civilians 25 individuals working in police laboratory	Switzerland	Hands	LC-MS	<ul style="list-style-type: none"> no positive civilian specimen two positive police specimens
Manganelli et al, 2019 [46]	OGSR	Civilian and police	122 civilians 115 police officers	Switzerland	Hands and wrists/sleeves	LC-MS	<ul style="list-style-type: none"> civilians: 18% of positive hand specimens and 11.5% wrists/sleeves police officers: 36.5% of positive hand specimens and 33% wrists/sleeves

79

DRAFT

80 The studies summarized in Table 1 show that prevalence can vary significantly depending on the
81 targeted population. The items/people directly in contact with firearms generally presented the highest
82 prevalence. Occupations involved in police forces or in firearm manufacture generally lead to a higher
83 background than for civilians. Similarly, activities such as hunting or recreational shooting should be
84 taken into account in the evaluation of OGSR evidence. To the best of our knowledge, only two studies
85 have investigated the presence of IGSR [34, 36] in police vehicles in North America. Both concluded
86 that the level of contamination was very low (one characteristic PbBaSb particle detected at most).
87 Another study investigated secondary transfer to volunteers from police vehicles, resulting in two
88 positive specimens, but did not take specimens from the vehicles themselves [44]. The number of studies
89 in vehicles remains very limited and no data regarding OGSR prevalence in police vehicles has been
90 published to date. The aim of the present study was thus to provide data pertaining to the OGSR
91 background levels of police vehicles in Switzerland. Specimens from 64 vehicles were collected from
92 two regional police services and analysed by LC-MS in positive mode. The driver's seat and the back
93 seats were sampled separately to monitor potential differences between locations and to assess the risks
94 of a suspect being contaminated by OGSR during transportation to a police station.

95

96 **2. MATERIALS AND METHODS**

97 **2.1 Specimen collection and preparation**

98 Specimens were collected from 64 police vehicles in collaboration with two regional police services.
99 Collection was performed using carbon stubs from Plano (Wetzlar, Germany), consisting of an adhesive
100 carbon tab 12 mm in diameter mounted on a 12.5 mm aluminium inserted in a plastic vial and sealed
101 with a screw cap. Two stubs were collected per vehicle: the first one from the driver's seat and the
102 second from the back seats. The stubs were dabbed about 200 times on the seats (the whole surface was
103 sampled), following recommendations from Zeichner *et al.* [47] for clothing items.

104

105 For compound extraction, the carbon adhesive was removed from the stub with clean tweezers and
106 transferred to a 20 mL scintillation vial containing 1 mL MeOH. The vial was placed in an ultrasonic
107 bath at room temperature for 15 minutes before filtration of the resulting extract through a 0.2 µm
108 Chromafil PTFE syringe filter (Macherey-Nagel, Düren, Germany) to remove carbon particles. To
109 detect potential laboratory contamination during specimen preparation, methanol blanks were prepared
110 before and after each extraction session. Likewise, a blank carbon tab was extracted to check for
111 potential contamination originating from the stub batch. For all these control samples, no OGSR were
112 detected.

113

114

115

116 2.3 Chemicals

117 Acetonitrile, methanol, formic acid (FA) and water were of ULC/MS grade (Biosolve, France). The
118 study targeted seven OGSR compounds: diphenylamine (DPA) from Fluka (Buchs, Switzerland);
119 ethylcentralite (EC), *N*-nitrosodiphenylamine (*N*-nDPA), 4-nitrodiphenylamine (4-nDPA), akardite II
120 (AK II) from Sigma–Aldrich (Buchs, Switzerland); 2-nitrodiphenylamine (2-nDPA) from Alfa Aesar
121 (Karlsruhe, Germany); methylcentralite (MC) from MP Biomedicals (Illkirch, France). Standard
122 solutions at 1 mg/mL were prepared in MeOH and stored at 4°C.

123

124 2.4 Instrumentation

125 The specimens were analysed using an Exion ultra-high performance liquid chromatography (UHPLC)
126 coupled to a QTrap 6500 from AB Sciex. The UHPLC instrument was equipped with a binary pump
127 enabling a maximum delivery flow rate of 10 mL/min, an autosampler, and a thermostatically-controlled
128 column compartment. Separation was obtained using a C18 Kinetex core-shell column (Phenomenex).
129 A C18 pre-column cartridge (SecurityGuard ULTRA) was placed before the analytical column for
130 protection. All UHPLC parameters are described in Table 2.

131

132 Table 2. UHPLC parameters

UHPLC parameters			
Column type	C18 (2.6 μ m, 2.1 mm \times 100 mm)		
Column temperature	40 °C		
Flow rate	0.4 mL/min		
Injection volume	5 μ L		
Gradient table	t / min	% A	% B
		<i>H₂O + 0.1% FA</i>	<i>ACN + 0.1% FA</i>
	0	65	35
	0.5	65	35
	4	40	60
	4.5	0	100
	5	0	100
	5.5	65	35
	7	65	35

133

134 Electrospray ionization was operated in positive mode. For all target compounds, the ion $[M+H]^+$ was
135 defined as the precursor ion and quantification was obtained from the SRM measurements (Table 3).
136 The source parameters were the following: the desolvation temperature (TEM) was set to 400°C, the
137 nebulizer gas to 65 psig, the turbo gas to 65 psig, the curtain gas to 30 psig and the IonSpray voltage to
138 5000 V. Data acquisition and instrument control were monitored using Analyst[®] software (version
139 1.6.3). Data treatment and quantitation were performed using MultiQuant[®] software (version 3.0.2).
140 Semi-quantitative data were obtained from a calibration curve (10 levels, 2 replicates) measured for each

141 sequence of experiments with levels ranging between the LOD and 10 ng/ml (EC and MC), 20 ng/mL
 142 (2-nDPA, 4-nDPA and AK II), 40 ng/mL (*N*-nDPA) and 100 ng/mL (DPA).

143 Table 3: MS parameters

Target compounds	SRM transitions	LOD [ng/mL]	Declustering potential [V]	Collision energy [V]	Collision cell exit potential [V]
Diphenylamine (DPA)	170.1 → 93.0 170.1 → 152.0	0.2	51 71	31 37	10 4
<i>N</i> -nitrosodiphenylamine (<i>N</i> -nDPA)	199.0 → 169.0 199.0 → 66.0	0.02	30 30	15 29	20 8
2-nitrodiphenylamine (2-nDPA)	215.0 → 180.0 215.0 → 198.0	0.02	60 60	23 19	20 20
4-nitrodiphenylamine (4-nDPA)	215.0 → 198.0 215.0 → 167.0	0.05	60 60	19 45	20 18
Ethylcentralite (EC)	269.1 → 148.0 269.1 → 120.0	0.005	80 80	19 29	16 14
Methylcentralite (MC)	241.1 → 134.0 241.1 → 106.0	0.005	60 60	20 33	16 14
Akardite II (AK II)	227.0 → 170.2 227.0 → 91.9	0.005	80 80	23 35	20 15

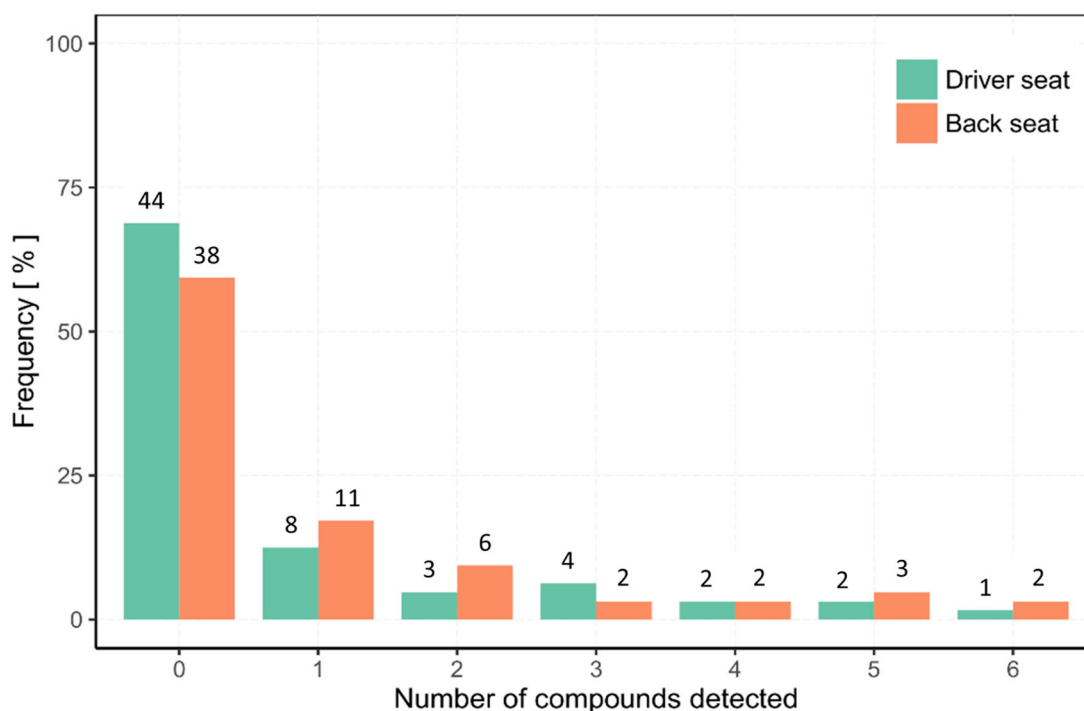
144

145 3. **RESULTS AND DISCUSSION**

146 The police vehicle population involved 64 vehicles, 31 from one regional police service and 33 from
 147 another. The vehicles had various functions: part were used by patrols for road surveillance or other
 148 community policing activities (Gendarmerie), others by the “Police de Sûreté”, and some for group
 149 and/or prisoner transportation. In the “Gendarmerie” whose main mission is to maintain security and
 150 order, the officers wear uniforms and generally drive marked police vehicles. The “Police de Sûreté” is
 151 mainly involved in investigations of crimes and officers are in civilian clothes. These officers generally
 152 travel in unmarked police cars. “Gendarmerie” vehicles often contain firearms that are permanently
 153 present within the vehicle and are rarely used in practice. The driver’s seat was targeted to evaluate the
 154 extent of secondary transfer from police officers. The back seat was sampled as well as it is used for
 155 suspect transportation and could be a contamination source. Data was collected regarding the permanent
 156 presence of one or more firearms and their location(s) within the vehicle, and whenever available the
 157 frequency of passenger compartment cleaning.

158 The results showed that most of the 64 vehicles were uncontaminated (44 driver’s seats and 38 back
 159 seats respectively, see Figure 1). Interestingly, the back seats (26 vehicles) were slightly more
 160 contaminated than the driver’s seats (20 vehicles). The number of compounds detected simultaneously
 161 was up to six out of the seven targeted, detected once on a driver’s seat and twice on back seats. As a
 162 general trend, increased numbers of compounds detected simultaneously exhibited lower detection
 163 frequencies. Thus, based on these data, the combined presence of four or more compounds in a police

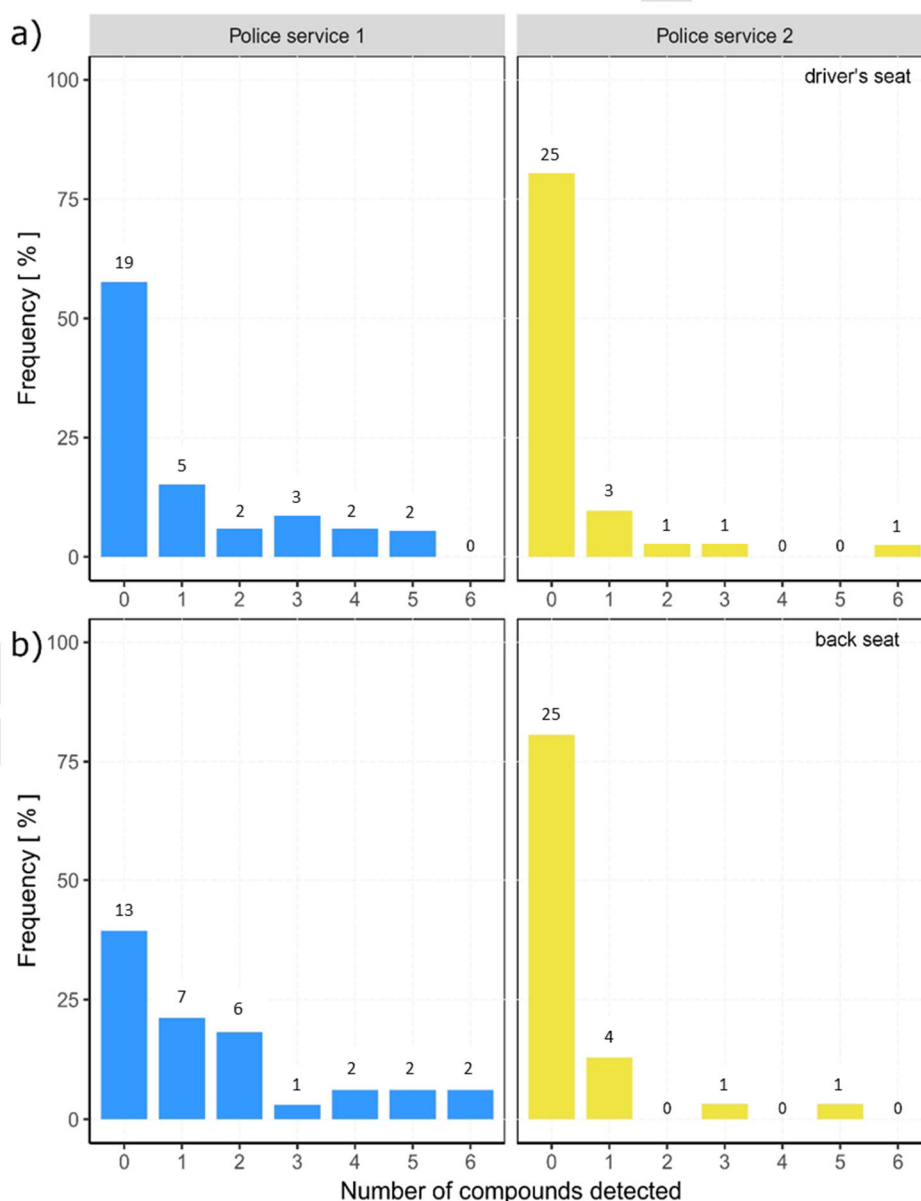
164 vehicle seat appears to be a relatively rare occurrence, as only nine cars presented such results (3 of them
165 both on the driver's and back seats).



166
167 Figure 1: Prevalence versus number of compounds detected in police vehicles (n = 64). The numbers of cars in each category
168 are indicated above the histogram bars.
169

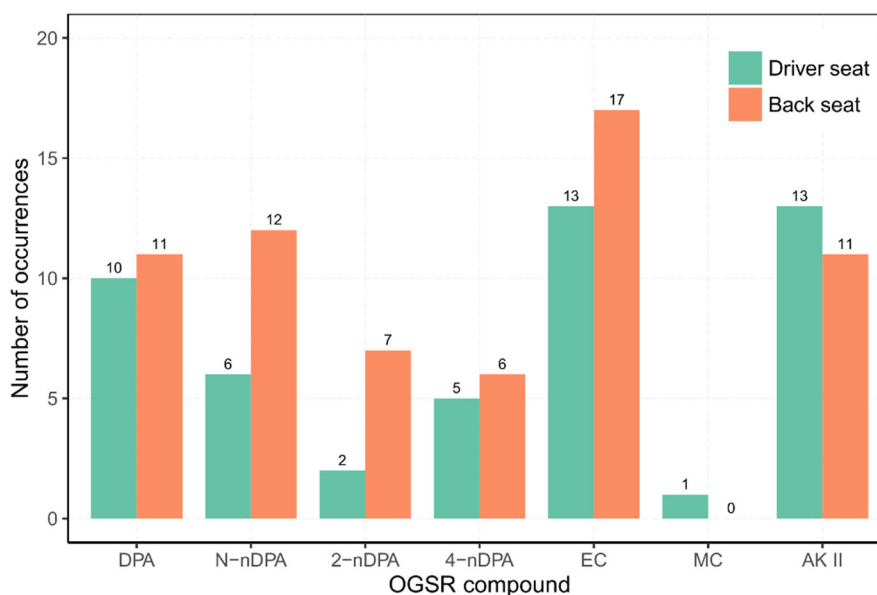
170 As the samples were collected from two regional police services, the data was separated according to its
171 origin to investigate potential differences (Figure 2). It can be observed that the OGSR background level
172 varied slightly between the two services. OGSR were detected in more cars from police service 1 (42.4%
173 and 60.6% for the driver and back seat respectively) than in cars from police service 2 (19.4% for both
174 driver and back seats). These results might be explained by three factors: the presence and location of
175 permanent firearms within the vehicle; the cleaning frequency; and the random sampling of cars
176 available at the time of specimen collection. In police service 1, the firearms (submachine guns) were
177 laid, covered by a blanket, over the back seat and/or in the trunk, whereas in police service 2, the guns
178 were stored in a box fastened to the car door or in a box attached to the rear of the back seat in the trunk.
179 The process of storing firearms in boxes might minimize seat contamination, as GSR might be deposited
180 inside the box instead of the seat. Such an explanation applies only to back seat contamination and
181 should not influence background levels on the driver's seat. One factor potentially influencing
182 background levels on both front and back seats is the cleaning frequency. Unfortunately, information
183 regarding cleaning frequency could not be obtained for all cars. For eleven cars in police service 1, the
184 last cleaning occurred five weeks before specimen collection, for one car three days and for another one
185 the day before. However, for the second service, marked police cars (representing about half of the
186 sampled vehicles) are cleaned (inside as well as outside) on a daily basis. For the unmarked cars, this

187 information could not be obtained. Interestingly, in police service 2, the unmarked cars had less OGSR
 188 background (two out of 17 cars with only one compound detected) than the marked cars that were
 189 cleaned on a daily basis (six out of 13). Major differences between those cars were their use by
 190 uniformed police officers, carrying a visible service weapon and the presence of guns within the car
 191 (marked cars) versus plainclothes police officers, carrying a concealed service weapon and the absence
 192 of other firearms within the vehicle. Such trend was not observed for police service 1 as the background
 193 levels were similar for marked and unmarked cars. However, it remains difficult to identify the real
 194 cause of the observed differences, as the limited number of cars sampled might not be representative of
 195 the whole vehicle population.



196
 197 Figure 2: Prevalence versus number of compounds detected on the a) driver seat, b) back seat of police vehicles as a function
 198 of police service ($n_1 = 33$, $n_2 = 31$). The numbers of cars in each category are indicated above the histogram bars.
 199

200 As to the number of times a specific OGSR compound was observed (Figure 3), EC was the most
 201 frequently encountered in the sampled vehicles (20% and 26.6% of all cars for the driver and back seats,
 202 respectively). AK II, DPA and *N*-nDPA followed with percentages between 9 and 20%. The DPA
 203 derivatives, 2-nDPA and 4-nDPA were slightly less frequently detected than DPA and *N*-nDPA. It is
 204 interesting to note that MC was only detected once. This might indicate that the propellant used by these
 205 police services in shooting training sessions or in duty does not contain this compound. No significant
 206 differences were observed in the occurrence of compounds between the two services.

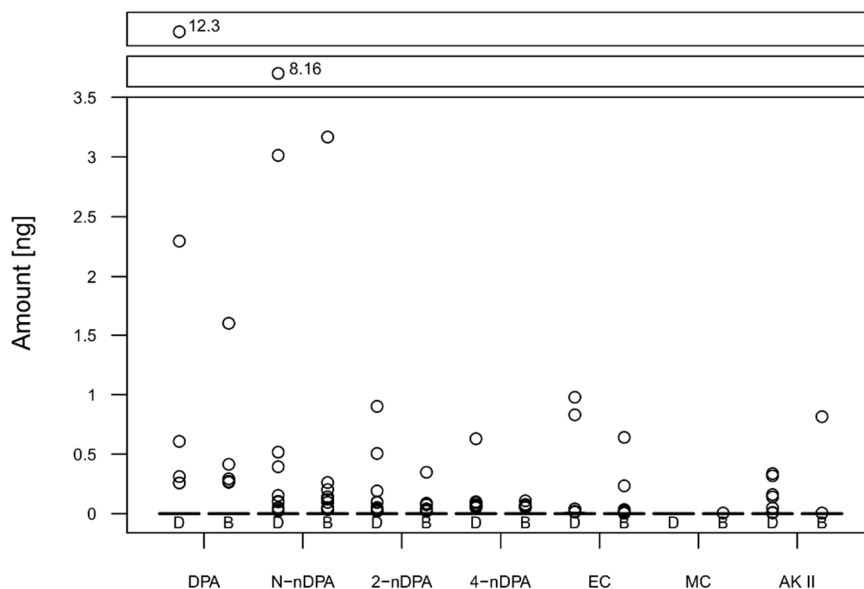


207
 208 Figure 3: Number of occurrences versus compound in police vehicles (n = 64)
 209

210 A recent study investigated OGSR prevalence on the hands and wrists/sleeves of police officers from
 211 three Swiss police services [46]. It indicated that *N*-nDPA and EC were the most frequently detected
 212 compounds (29.6% and 21.7% on hands for *N*-nDPA and EC respectively), closely followed by DPA
 213 plus derivatives and AK II. MC was also only detected once. In spite of the different percentages, the
 214 results between both studies regarding the type of compounds detected are in good agreement. Thus, the
 215 OGSR compounds detected in police vehicles are most certainly transferred from police officers and
 216 firearms through secondary transfer. Nevertheless, an environmental source cannot be totally excluded
 217 when for example only one compound is detected. A background study involving new vehicles could
 218 provide information on the environmental presence of these compounds on car seats. However, the
 219 absence of OGSR compounds in most of the police vehicles tends to indicate that these compounds are
 220 not normally present on car seats.

221
 222 The amounts detected in the specimens (Figure 4) were in the low and sub-nanogram range, except for
 223 two specimens at 12.3 ng/mL (DPA) and 8.16 ng/mL (*N*-nDPA). It is interesting to see that the highest
 224 values were for DPA and *N*-nDPA, while the other compounds were all detected below 1 ng/mL. There
 225 were no significant differences between the amounts detected on the driver's and back seats. The results

226 of Figure 4 were represented as boxplots, but only outliers were observed, as the values for the medians,
 227 the first and third quartiles were all equal to zero. This highlights the low number of cars positive to
 228 OGSR and the very low amounts detected, thus indicating that detecting a specific compound happened
 229 in less than 25% of the specimens. Compared to amounts that are detected on the hands of shooters just
 230 after discharge or to the prevalence study of police officers involving sampling just after shooting
 231 training (highest values above 100 ng), the amounts detected in vehicles are much lower.



232
 233 Figure 4: Prevalence in the police vehicle population: Amount of analyte detected. The letters D and B denote the driver and
 234 back seat respectively (n = 64)
 235

236 The specimens with the highest compound numbers (five and six) were investigated in greater depth in
 237 order to explain the background level and to qualitatively evaluate if there was a correlation between
 238 the number of compounds and the amount detected on both seats. In total, six vehicles had five or six
 239 compounds detected on either the driver or the back seat (Table 4). While in four vehicles, contamination
 240 was present on both seats, it was not the case for two cars. In vehicles 20 and 29, no OGSR was detected
 241 on the driver's seat, whereas six and five compounds were detected on the back seat respectively. Thus,
 242 detecting a high number of compounds on the back seat was not indicative of similar contamination of
 243 the driver's seat. It is thus reasonable to assume that contamination of driver and back seats is not
 244 necessarily correlated and that secondary transfer may occur independently from two separate sources.

245
 246
 247
 248
 249
 250
 251

252 Table 4: summary of the results from the six police vehicles with the highest background level

Car	Driver seat OGSR		Back seat OGSR nb	
	Number of compounds	Compounds detected	Number of compounds	Compounds detected
Car 16	5	<i>N</i> -nDPA, 4-nDPA, 2-nDPA, AK II, EC	4	<i>N</i> -nDPA, 2-nDPA, AK II, EC
Car 20	0	-	6	DPA, <i>N</i> -nDPA, 4-nDPA, 2-nDPA, AK II, EC
Car 26	2	<i>N</i> -nDPA, EC	6	DPA, <i>N</i> -nDPA, 4-nDPA, 2-nDPA, AK II, EC
Car 29	0	-	5	DPA, <i>N</i> -nDPA, 4-nDPA, 2-nDPA, EC
Car 33	5	DPA, <i>N</i> -nDPA, 4-nDPA, 2-nDPA, EC	5	DPA, <i>N</i> -nDPA, 2-nDPA, AK II, EC
Car 4	6	DPA, <i>N</i> -nDPA, 4-nDPA, 2-nDPA, AK II, EC	5	DPA, <i>N</i> -nDPA, 4-nDPA, 2-nDPA, EC

253
 254 For car 16, a police officer had used the vehicle to go to a shooting session two months before specimen
 255 collection. There was no firearm present permanently in the car. No information regarding the last
 256 cleaning was available. Contamination through secondary transfer from the shooting police officer
 257 might be a valid hypothesis as OGSR persistence is expected to be longer on car seats compared with
 258 hands. However, no data is currently available and the persistence of such contamination should be
 259 investigated. For car 20, no firearm was stored in the vehicle, but it was used to drive groups of police
 260 officers to courses or shooting sessions, which might explain the presence of OGSR on the back seats.
 261 No information regarding last cleaning could be obtained. For car 29, a submachine gun was present on
 262 the back seat, but no information could be obtained regarding its recent use or manipulation. Contact
 263 with the submachine gun might be an explanation for that contamination. For car 4, a gun was stored in
 264 a box fastened to a door and the car was theoretically cleaned in the last 24 hours, so it is more difficult
 265 to find an explanation for this background. Can a contamination persist in spite of the cleaning or was
 266 it contaminated after the cleaning? It must be highlighted that for each compound, car 4 had the highest
 267 values detected in the study. For cars 26 and 33, no firearm was present in the car, no information
 268 regarding last cleaning was available and no concrete explanation could be found. In all cases, vehicles
 269 were used by police officers carrying firearms and these might be a source of contamination, even though
 270 the present results indicate that it is a relatively rare occurrence.

271
 272 In terms of degree of contamination, the scenario producing the highest amounts and numbers of OGSR
 273 on car seats would probably be the discharge of a firearm within the vehicle. That would represent a
 274 primary transfer scenario. The use of a firearm on duty is anecdotal in Switzerland. The discharge of a
 275 firearm within a police vehicle would be even rarer. No data regarding the amounts to be expected in
 276 such an instance was found in the literature and it would be interesting to perform some experiments to
 277 assess the degree of contamination that can result from a discharge within a vehicle. Experiments

278 performed by Burnett and Lebiezick for IGSR showed heavy contamination of interior surfaces such as
279 the dashboard or the window frame when a firearm was discharged by the driver of a vehicle through
280 an opened window [32]. Secondary transfer scenarios would most probably lead to lower background
281 levels than primary transfer scenarios as evidenced by some studies regarding secondary transfer from
282 a shooter or a firearm to a third party [29, 30]. The most common hypotheses would then be secondary
283 transfer from contaminated police officers or from firearms that are either present or manipulated in the
284 vehicle. The background level of police vehicles might also be influenced by factors such as the car
285 cleaning frequency, the number of users and frequency of use. One might expect a higher OGSR
286 contamination if a vehicle is used to go to a shooting training than during normal duty. The same line of
287 reasoning holds for the number of users, as the probability of secondary transfer might increase with
288 that number. Logically, the more frequently a car is used, the higher the probability of secondary transfer
289 from the various users. However, it is also possible that the persistence of OGSR might decrease when
290 the number of users and the frequency of the vehicle use increases. Such parameters might be interesting
291 study perspectives.

292
293 Other OGSR prevalence studies involving cars could not be found in the literature. However, two studies
294 investigated the presence of IGSR in police vehicles. Berk et al. found one PbBaSb particle in two
295 vehicles [34] and Gerard et al. found one PbBaSb particle in two of the 18 vehicles sampled [36]. Both
296 studies concluded that the risk of secondary transfer from police vehicles was low, even though possible.
297 The present study for OGSR showed that most of the police vehicles sampled were free from OGSR.
298 However, up to six compounds were detected simultaneously in a number of vehicles in amounts up to
299 10 ng. As a precaution, suspect transportation should be performed in cars not used by heavily
300 contaminated users or within which a firearm is present or was manipulated. Another recommendation
301 would be the regular cleaning (vacuuming) of the vehicle's interior surfaces with a monitoring of the
302 efficiency of the procedure on removal of GSR in general. Experiments involving the transportation of
303 individuals in police vehicles would provide data as to the real risk of OGSR tertiary transfer from the
304 seats to an individual transported in such vehicle.

305 306 **4. CONCLUSIONS**

307
308 The present study aimed at evaluating the OGSR background level in police vehicles. Specimens from
309 the driver and back seats were separately collected from 64 cars from two regional police services in
310 Switzerland. The results showed that most of the 64 vehicles were uncontaminated (44 driver seats and
311 38 back seats respectively). The number of compounds detected on a single seat was up to six
312 compounds, detected once on a driver and twice on back seats. A trend was observed, as the
313 contamination frequency decreased with the number of compounds detected together. The amounts
314 detected were in the low ng range and inferior to amounts generally detected just after discharge on a

315 shooter. Our data indicate that detecting a combination of four or more compounds on a police vehicle
316 seat appears to be a relatively rare occurrence.

317 In the light of the anecdotal firearm use on duty in Switzerland, it seems logical that the background
318 contamination observed is due to secondary transfer from police officers (for example contaminated
319 through recent participation to a shooting session or firearm manipulation) or from firearms stored in
320 the vehicles. Thus, the background might be different in other countries in which firearms are more
321 often used on duty (in a car chase for example). The present results might be used as a recommendation
322 to minimize contact of a suspect with contaminated surfaces if OGSR is implemented in routine work
323 in parallel to IGSR analysis. Therefore, regular cleaning of police vehicles' interior surfaces and
324 monitoring of the GSR background of cars usually used for suspect transportation should be performed.
325 Moreover, to help interpretation of the GSR evidence, a specimen from the vehicle might also be
326 collected before suspect transportation as a blank to evaluate risks of secondary transfer from the police
327 car back seats. Another option would be to protect the hands of the suspect by plastic bags. However,
328 that would not exclude potential contamination from the car seats to the suspect's clothing for example.

330 **Acknowledgements**

331 The authors wish to acknowledge the Swiss National Science Foundation (10521A_165608) for
332 financial support. They also would like to thank two Swiss police services for enabling the collection of
333 the vehicle prevalence specimens, especially Commissaire Nicola Albertini, Adjudant Jean-Philippe
334 Jaquier, Inspecteur Arnaud Yersin from the Police Cantonale Vaudoise, Adjudant Dr Balthasar Jung,
335 Sergeant Manuela Manganelli from the Kantonspolizei Aargau. They are grateful to Dr Amanda Frick
336 for proof reading this manuscript, to Virginie Redouté Minzière for helping with part of the specimen
337 collection process and to Dr Ana Moraleda Merlo for preparation of the calibration standards.

339 **Bibliography**

- 340
- 341 [1] Zeichner A. Recent developments in methods of chemical analysis in investigations of firearm-related events.
342 *Anal Bioanal Chem.* 2003;376:1178-91.
- 343 [2] Wallace JS. *Chemical Analysis of Firearms, Ammunition, and Gunshot Residue.* Boca Raton: CRC Press;
344 2008.
- 345 [3] Wolten GM, Nesbitt RS, Calloway AR, Loper GL, Jones PF. *Equipment Systems Improvement Program: Final*
346 *Report on Particle Analysis for Gunshot Residue Detection.* Law Enforcement Development Group. USA: The
347 *Aerospace Corporation;* 1977.
- 348 [4] Zeichner A. *Firearm Discharge Residue: Analysis of.* In: John Wiley & Sons L, editor. *Wiley Encyclopedia of*
349 *Forensic Science*2009.
- 350 [5] Meng HH, Caddy B. *Gunshot residue analysis - A review.* *J Forensic Sci.* 1997;42:553-70.
- 351 [6] Dalby O, Butler D, Birkett JW. *Analysis of Gunshot Residue and Associated Materials-A Review.* *J Forensic*
352 *Sci.* 2010;55:924-43.
- 353 [7] Lopez-Lopez M, Delgado JJ, Garcia-Ruiz C. *Ammunition Identification by Means of the Organic Analysis of*
354 *Gunshot Residues Using Raman Spectroscopy.* *Anal Chem.* 2012;84:3581-5.
- 355 [8] Abrego Z, Grijalba N, Unceta N, Maguregui M, Sanchez A, Fernandez-Isla A, et al. *A novel method for the*
356 *identification of inorganic and organic gunshot residue particles of lead-free ammunitions from the hands of*
357 *shooters using scanning laser ablation-ICPMS and Raman micro-spectroscopy.* *Analyst.* 2014;139:6232-41.

358 [9] Bueno J, Lednev IK. Raman microspectroscopic chemical mapping and chemometric classification for the
359 identification of gunshot residue on adhesive tape. *Anal Bioanal Chem.* 2014;406:4595-9.
360 [10] Bueno J, Sikirzhyski V, Lednev IK. Attenuated Total Reflectance-FT-IR Spectroscopy for Gunshot Residue
361 Analysis: Potential for Ammunition Determination. *Anal Chem.* 2013;85:7287-94.
362 [11] Bueno J, Lednev IK. Attenuated Total Reflectance-FT-IR Imaging for Rapid and Automated Detection of
363 Gunshot Residue. *Anal Chem.* 2014;86:3389-96.
364 [12] Arndt J, Bell S, Crookshanks L, Lovejoy M, Oleska C, Tulley T, et al. Preliminary evaluation of the
365 persistence of organic gunshot residue. *Forensic Sci Int.* 2012;222:137-45.
366 [13] Moran JW, Bell S. Skin Permeation of Organic Gunshot Residue: Implications for Sampling and Analysis.
367 *Anal Chem.* 2014;86:6071-9.
368 [14] Yeager B, Bustin K, Stewart J, Dross R, Bell S. Evaluation and validation of ion mobility spectrometry for
369 presumptive testing targeting the organic constituents of firearms discharge residue. *Anal Methods-Uk.*
370 2015;7:9683-91.
371 [15] Northrop DM. Gunshot residue analysis by micellar electrokinetic capillary electrophoresis: Assessment for
372 application to casework. Part I. *J Forensic Sci.* 2001;46:549-59.
373 [16] Northrop DM. Gunshot residue analysis by micellar electrokinetic capillary electrophoresis: Assessment for
374 application to casework. Part II. *J Forensic Sci.* 2001;46:560-72.
375 [17] Reardon MR, MacCrehan WA, Rowe WF. Comparing the additive composition of smokeless gunpowder and
376 its handgun-fired residues. *J Forensic Sci.* 2000;45:1232-8.
377 [18] MacCrehan WA, Patierno ER, Duewer DL, Reardon MR. Investigating the effect of changing ammunition
378 on the composition of organic additives in gunshot residue (OGSR). *J Forensic Sci.* 2001;46:57-62.
379 [19] Muller D, Levy A, Vinokurov A, Ravreby M, Shelef R, Wolf E, et al. A novel method for the analysis of
380 discharged smokeless powder residues. *J Forensic Sci.* 2007;52:75-8.
381 [20] Zeichner A, Eldar B. A novel method for extraction and analysis of gunpowder residues on double-side
382 adhesive coated stubs. *J Forensic Sci.* 2004;49:1194-206.
383 [21] Tarifa A, Almirall JR. Fast detection and characterization of organic and inorganic gunshot residues on the
384 hands of suspects by CMV-GC-MS and LIBS. *Sci Justice.* 2015;55:168-75.
385 [22] Laza D, Nys B, De Kinder J, Mesmaeker AKD, Moucheron C. Development of a quantitative LC-MS/MS
386 method for the analysis of common propellant powder stabilizers in gunshot residue. *J Forensic Sci.* 2007;52:842-
387 50.
388 [23] Thomas JL, Lincoln D, McCord BR. Separation and Detection of Smokeless Powder Additives by Ultra
389 Performance Liquid Chromatography with Tandem Mass Spectrometry (UPLC/MS/MS). *J Forensic Sci.*
390 2013;58:609-15.
391 [24] Benito S, Abrego Z, Sanchez A, Unceta N, Goicolea MA, Barrio RJ. Characterization of organic gunshot
392 residues in lead-free ammunition using a new sample collection device for liquid chromatography-quadrupole
393 time-of-flight mass spectrometry. *Forensic Sci Int.* 2015;246:79-85.
394 [25] Taudte RV, Roux C, Blanes L, Horder M, Kirkbride KP, Beavis A. The development and comparison of
395 collection techniques for inorganic and organic gunshot residues. *Anal Bioanal Chem.* 2016;408:2567-76.
396 [26] Gassner AL, Weyermann C. LC-MS method development and comparison of sampling materials for the
397 analysis of organic gunshot residues. *Forensic Sci Int.* 2016;264:47-55.
398 [27] BKA. Development of analytical methods for sensitive detection and identification of organic gunshot
399 residues (OGSR) based on liquid chromatography-mass spectrometry (LC-MS) for routine casework. 2017.
400 [28] Maitre M, Horder M, Kirkbride KP, Gassner AL, Weyermann C, Roux C, et al. A forensic investigation on
401 the persistence of organic gunshot residues. *Forensic Sci Int.* 2018;292:1-10.
402 [29] Gassner AL, Manganelli M, Werner D, Rhumorbarbe D, Maitre M, Beavis A, et al. Secondary transfer of
403 organic gunshot residues: Empirical data to assist the evaluation of three scenarios. *Sci Justice.* 2019;59:58-66.
404 [30] Maitre M, Chadwick S, Kirkbride PK, Gassner AL, Weyermann C, Beavis A, et al. An investigation on the
405 secondary transfer of organic gunshot residues. *Sci Justice.* 2019;In Press.
406 [31] Romolo FS, Margot P. Identification of gunshot residue: a critical review. *Forensic Sci Int.* 2001;119:195-
407 211.
408 [32] Burnett BR, Lebidzik J. Discharge of a Pistol Out a Car Window with the Breech Within the Interior of the
409 Car: Analysis of Gunshot Residue on a Car's Interior Surfaces. *J Forensic Sci.* 2017;62:768-72.
410 [33] Gialamas DM, Rhodes EF, Sugarman LA. Officers, Their Weapons and Their Hands - an Empirical-Study of
411 Gsr on the Hands of Non-Shooting Police Officers. *J Forensic Sci.* 1995;40:1086-9.
412 [34] Berk RE, Rochowicz SA, Wong M, Kopina MA. Gunshot residue in Chicago police vehicles and facilities:
413 An empirical study. *J Forensic Sci.* 2007;52:838-41.
414 [35] Lindsay E, McVicar MJ, Robert V. Gerard RV, Randall ED. Observations of GSR on the Hands of Employees
415 at Firearms Manufacturing Facilities. *Canadian Society of Forensic Science Journal.* 2011;44:105-9.

- 416 [36] Gerard RV, Lindsay E, McVicar MJ, Randall ED, Gapinska A. Observations of Gunshot Residue Associated
417 with Police Officers, Their Equipment, and Their Vehicles. *Canadian Society of Forensic Science Journal*.
418 2012;45:57-63.
- 419 [37] Brozek-Mucha Z. On the prevalence of gunshot residue in selected populations - An empirical study
420 performed with SEM-EDX analysis. *Forensic Sci Int*. 2014;237:46-52.
- 421 [38] Hannigan TJ, McDermott SD, Greaney CM, O'Shaughnessy J, O'Brien CM. Evaluation of gunshot residue
422 (GSR) evidence: Surveys of prevalence of GSR on clothing and frequency of residue types. *Forensic Sci Int*.
423 2015;257:177-81.
- 424 [39] Cook M. Gunshot residue contamination of the hands of police officers following start-of-shift handling of
425 their firearm. *Forensic Sci Int*. 2016;269:56-62.
- 426 [40] Lucas N, Brown H, Cook M, Redman K, Condon T, Wrobel H, et al. A study into the distribution of gunshot
427 residue particles in the random population. *Forensic Sci Int*. 2016;262:150-5.
- 428 [41] Comanescu MA, Millett TJ, Kubic TA. A Study of Background Levels of Antimony, Barium, and Lead on
429 Vehicle Surface Samples by Graphite Furnace Atomic Absorption. *J Forensic Sci*. 2019;64:565-9.
- 430 [42] Lucas N, Cook M, Kirkbride KP, Kobus H. Gunshot residue background on police officers: Considerations
431 for secondary transfer in GSR evidence evaluation. *Forensic Sci Int*. 2019;297:293-301.
- 432 [43] Bell S, Seitzinger L. From binary presumptive assays to probabilistic assessments: Differentiation of shooters
433 from non-shooters using IMS, OGSR, neural networks, and likelihood ratios. *Forensic Sci Int*. 2016;263:176-85.
- 434 [44] Ali L, Brown K, Castellano H, Wetzel SJ. A Study of the Presence of Gunshot Residue in Pittsburgh Police
435 Stations using SEM/EDS and LC-MS/MS. *J Forensic Sci*. 2016;61:928-38.
- 436 [45] Hofstetter C, Maitre M, Beavis A, Roux CP, Weyermann C, Gassner AL. A study of transfer and prevalence
437 of organic gunshot residues. *Forensic Sci Int*. 2017;277:241-51.
- 438 [46] Manganelli M, Weyermann C, Gassner AL. Surveys of organic gunshot residue prevalence: Comparison
439 between civilian and police populations. *Forensic Sci Int*. 2019;298:48-57.
- 440 [47] Zeichner A, Levin N. Collection Efficiency of Gunshot Residue (Gsr) Particles from Hair and Hands Using
441 Double-Side Adhesive Tape. *J Forensic Sci*. 1993;38:571-84.

442